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A high flow rate, very low pressure drop impactor for inertial separation of ultrafine from accumulation mode particles

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Abstract

This paper presents the development and evaluation of a high-volume, multiple rectangular (slit) geometry jet impactor. Operating with a preselective inlet that removes particles larger than $2.5\ \mu\text{m}$ in aerodynamic size, the impactor has been designed to separate ultrafine ($< 0.15\ \mu\text{m}$) from the accumulation mode range ($0.15 < d_p < 2.5\ \mu\text{m}$). Particles are accelerated by passing through 10 parallel slits nozzles, each $12.5\ \text{cm}$ long by $0.0125\ \text{cm}$ wide. The average jet velocity in each rectangular jet is approximately $5800\ \text{cm s}^{-1}$. Following acceleration, particles larger than approximately $0.15\ \mu\text{m}$ impact on quartz fiber strips, each $12.5 \times 0.5\ \text{cm}^2$, while the aerosol fraction smaller than $0.15\ \mu\text{m}$ penetrates through the outlet of the impactor. The impactor operates at a flow rate $550\ \text{l min}^{-1}$ and at a very low pressure drop of $0.020\ \text{kPa}$. The performance of the multi-slit impactor was validated in laboratory and the field tests. Laboratory experiments conducted with monodisperse PSL particles as well as polydisperse ammonium nitrate, sulfate and indoor aerosols corroborated the 50% cutpoint of the impactor at $0.15\ \mu\text{m}$. Field test comparisons between the high-volume multi-slit impactor and the Microorifice Uniform Deposit Impactor (MOUDI) showed that the accumulation and ultrafine mode concentrations of particulate nitrate, sulfate, elemental and organic carbon are in very good agreement (within 10% or less). This impactor has been developed primarily as a separator of ultrafine from accumulation mode particles for use in human exposure studies to concentrated ambient ultrafine aerosols. High-volume collection of size-fractionated particulate matter accomplished by this impactor further enables investigators in the field of environmental health to conduct toxicological studies using ambient accumulation and ultrafine mode particles in vitro as well as by means of intratracheal instillation. © 2002 Elsevier Science Ltd. All rights reserved.

Keywords: Inertial impaction; Slit nozzle; Accumulation mode and ultrafine particles; Impaction substrates

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1. Introduction

There is abundant epidemiological evidence associating increased air particulate pollution and the incidence of adverse health effects on humans (Dockery, Speizer, Stram, Ware, Spengler, & Ferris, 1989; Schwartz & Dockery, 1992). These associations have been primarily demonstrated for fine particles (e.g. particles smaller than $2.5\text{ }\mu\text{m}$ in size, also referred to as $\text{PM}_{2.5}$) and their components such as sulfate (SO_4^{2-}) and strong acidity (H^+) (Bates & Sizto, 1989; Dockery & Pope, 1994). Although epidemiological studies to date have not made it perfectly clear whether it is particle mass, surface area or number concentrations that may be responsible for these observed health outcomes presumably attributable to particulate matter (PM), certain toxicological investigations suggest that atmospheric ultrafine particles may be responsible for some of these adverse effects (Oberdorster, Ferin, Gelein, Soderholm, & Finkelstein, 1992; Oberdorster, Gelein, Ferin, & Weiss, 1995; Dreher, Jaskot, Lehmann, Richards, McGee, Ghio, & Costa, 1997; Donaldson, Li, & MacNee, 1998).

Atmospheric ultrafine particles arise from either gas-to-particle conversion processes, in which hot, supersaturated vapors undergo condensation upon being cooled to ambient temperatures and/or are directly emitted as products of incomplete combustion processes (Whitby & Svendrup, 1980; Finlayson-Pitts & Pitts, 1986). Particles in the approximate size range of $0.01\text{--}0.15\text{ }\mu\text{m}$ are formed entirely during these processes. Although the mass fraction of the ultrafine mode is negligible, this size range contains the highest number of ambient particles by counts as well as the surface area. Because of their increased number and surface area, ultrafine particles are particularly important in atmospheric chemistry and environmental health.

To date there has been limited, but rapidly increasing toxicological and epidemiological evidence linking respiratory health effects and exposures to ultrafine particles. The limited data are primarily due to lack of adequate methods for ultrafine particle separation and measurement. Nevertheless, recent epidemiological studies (Heyder, Brand, Heinrich, Peters, Scheuh, Tuch, & Wichmann, 1996; Peters, Dockery, Heinrich, & Wichmann, 1997), demonstrated a stronger association between health effects and exposures to ultrafine particles compared to accumulation or coarse particles. Toxicological studies by Donaldson et al. (1998) and Oberdorster et al. (1992) indicated that ultrafine particles exerted a stronger physiological effect than the same mass of coarse or fine particles. More recently, a study by Dailey, Kim, Sioutas, and Devlin (2001) indicated that, when airway human epithelial cells are exposed on an equal mass basis to the different PM fractions, cells exposed to ultrafine particles demonstrate a greater response at a lower mass concentrations compared to accumulation or coarse mode particles.

Inhaled ultrafine particles deposit primarily in the lower respiratory tract and alveoli by diffusional mechanisms (ICRP, 1994). The pulmonary toxicity of ultrafine particles has been demonstrated in several controlled laboratory exposure studies. Oberdorster et al. (1992) have shown that apparently “inert” dusts consisting of ultrafine particles can be highly toxic to the lung because of the interstitial access across the alveolar epithelium where they interact with macrophages. It is conceivable that when subjected to a high ultrafine particle number concentration, “overload” may occur, in which the alveolar macrophages themselves cause injury to the lungs by releasing excessive quantities of oxygen radicals, proteases, and certain proteins (Oberdorster et al., 1992). Other investigators have suggested that ultrafine particles coated with neutralized strong acids may, upon deposition, cause tissue damage due to their acidity (Ferin, Oberdorster, Soderholm, & Gelein, 1991). In addition, acids or catalytic metals on the surfaces of irritant particles can be more efficiently transferred to

the lungs by ultrafines than by an equal mass of larger particles. This is both because of the larger surface area-to-mass ratio inherent in the smaller particles and because the ultrafines have a higher deposition efficiency in the alveolar region. Finally, investigators have suggested that trace metals distributed widely throughout the lung on ultrafine particles could catalyze the formation of oxidants within pollutant concentrations in relation to health statistics within the lung which in turn produce tissue damage (Ghio et al., 1997; Dreher et al., 1997).

If these distinctions attributed to ultrafine particles are proven to be physiologically important, they may help to explain the difference between epidemiological findings inferred from exposure to real ambient particles versus those laboratory studies that in the past have been based largely on laboratory-generated aerosols. The few inhalation experiments that have been conducted to date using ultrafine particles have been conducted with model Teflon and TiO_2 aerosols (Oberdorster et al., 1992; Ferin, Oberdorster, & Penny, 1992; Donaldson, Stone, Gilmour, Brown, & MacNee, 2000). Studies have also been conducted using combustion generated nickel species aerosol, NiO and NiSO_4 to assess lung injury (Miller, Biswas, & Leikauf, 2001). The recent development of fine plus ultrafine particle concentrators based on the principle of virtual impaction (Kim, Chang, Kim, & Sioutas, 2000; Kim, Jaques, Chang, & Sioutas, 2001a, b) has made it possible to perform controlled exposures with “real-life” ambient aerosols at highly increased, but still environmentally realistic concentrations. This new line of investigation may eventually lead to coherence between laboratory studies and epidemiological evidence. Compared to previously developed technologies used to concentrate mainly the accumulation PM mode (Sioutas, Koutrakis, Ferguson, & Burton, 1995; Sioutas, Koutrakis, Godleski, Ferguson, Kim, & Burton, 1997), these newly developed concentrators have the ability to enrich the concentrations of fine particles ($0.01\text{--}2.5\text{ }\mu\text{m}$) including their ultrafine component. In order to assess solely the toxicity of ambient atmospheric ultrafine particles using these new concentrators, ultrafine particles need to be separated from the overwhelmingly larger mass of the accumulation mode of $\text{PM}_{2.5}$. Ideally, this separation should also occur under near-atmospheric pressure conditions in order to make human or animal exposures to these particles feasible.

Separation of ultrafine particles from the air sample by inertial impaction is a technical challenge. Hypersonic impactors operating at very low pressures (typically on the order of 500 Pa or less) have been developed to sample ultrafine particles (Fernandez de la Mora, Hering, Rao, & McCurry, 1990; Hering & Stolzenburg, 1995; Olawoyin, Raunemaa, & Hopke, 1995). Use of hypersonic impactors to separate ultrafine particles from the air sample, however, can be problematic because the separated particles are at a very low pressure, which makes them impossible to be used in inhalation exposures.

This paper discusses the development and characterization of multiple rectangular (slit) geometry jet conventional impactor that can be used to separate ultrafine from accumulation mode particles at high flow rates (550 l min^{-1}) and under a very low pressure drop (i.e., 8 in. of H_2O or 0.020 kPa). These two features are extremely important in making human exposures to concentrated atmospheric ultrafine aerosols possible, as these exposures require that the concentrated aerosol is supplied to the exposure chamber at a much higher flow than the typical human breathing rate ($12\text{--}30\text{ l min}^{-1}$) and at near-atmospheric pressure. The key feature of this technique is the utilization of a multiple slit nozzle conventional impactor with a small 50% cutpoint ($0.15\text{ }\mu\text{m}$) to sample atmospheric particulate pollutants at high flow rates. Particles larger than $0.15\text{ }\mu\text{m}$ in diameter are collected on narrow strips ($12.5\text{ cm} \times 0.254\text{ cm}$), which serve as the impaction substrate.

This impactor has been developed primarily as a separator of ultrafine from accumulation mode particles for use in human exposure studies to concentrated ambient ultrafine aerosols, by removing

the accumulation PM from the ambient air sample prior to concentration enrichment. Ultrafine particles penetrating the impactor can either be directly supplied to exposure chambers, or collected on standard, commercially available, $20 \times 25.4 \text{ cm}^2$ quartz or Teflon filters. High-volume collection of size-fractionated PM accomplished by this impactor further enables investigators in the field of environmental health to conduct toxicological studies using ambient accumulation and ultrafine mode particles in vitro as well as by means of intratracheal instillation.

2. Experimental methods

2.1. Impactor design

The ultrafine high-volume, low cutpoint impactor, shown in Figs. 1a–c, is a very compact and light weight (2.3 kg) conventional impactor made of aluminum. Its outside dimensions are $17 \text{ cm} \times 17 \text{ cm} \times 2.54 \text{ cm}$. Because of its very low pressure drop (0.020 kPa) the impactor can be operated using a relatively small pump (GAST 2067, 0.75 kW, Gast Corp., Benton Harbor, MI) operating at 110 V, single phase, thus does not require a large blower or a three-phase circuit. This greatly enhances its versatility for use in various field locations, in which reduction in the overall power required for air sampling is a highly desirable feature. If removal of the coarse PM mode (i.e., $2.5\text{--}10 \text{ }\mu\text{m}$) is desired prior to separation of the accumulation from the ultrafine modes, the impactor's inlet has been made such that it can be connected directly to a commercially available high-volume virtual impactor (HVVI Model 340, MSP Corp., Minneapolis, MN) developed by Marple, Liu, and Burton (1990). The HVVI is a multi-nozzle round jet virtual impactor, operating at 1130 l min^{-1} (hence twice the flow of the USC ultrafine separator) and having a 50% cutpoint at $2.5 \text{ }\mu\text{m}$ with a 5% minor-to-total flow ratio. To couple it with the USC ultrafine-accumulation PM separator, half of the round-nozzle virtual impactation jets of HVVI are masked for it to operate at 550 l min^{-1} .

The impactor consists of four different parts as shown in Fig. 1. The inlet block (Fig. 1a) has a circular opening, 5.08 cm in inside diameter. After entering through the inlet, the aerosol stream is accelerated by passing through the 10 parallel slit nozzles, each 12.5 cm long by 0.0125 cm wide. The average jet velocity in each rectangular jet is approximately 5800 cm s^{-1} . Following acceleration, particles larger than approximately $0.15 \text{ }\mu\text{m}$ impact upon the collection substrates on the collecting blocks, while the aerosol fraction smaller than $0.15 \text{ }\mu\text{m}$ penetrates through the outlet of the impactor.

The choice of the sampling flows as well as the nozzle design parameters was made so that the predicted cutpoint of the impactor will be about $0.15 \text{ }\mu\text{m}$. The principal parameter determining particle capture is the Stokes number of a particle having a 50% probability of impacting, St , defined the ratio of the product of the jet velocity, U , and the particle relaxation time, τ , versus the impactor's nozzle diameter (round nozzle) or width (rectangular nozzle such as the USC ultrafine impactor), W (Hinds, 1982):

$$St = \frac{\tau U}{W} = \frac{\rho_p C_c d_p^2 U}{9\mu W}, \quad (1)$$

where W is the diameter of the impactor's nozzle, U is the average velocity of the impactor jet, ρ_p is the particle density, μ is the dynamic viscosity of the air and C_c is the Cunningham slip correction

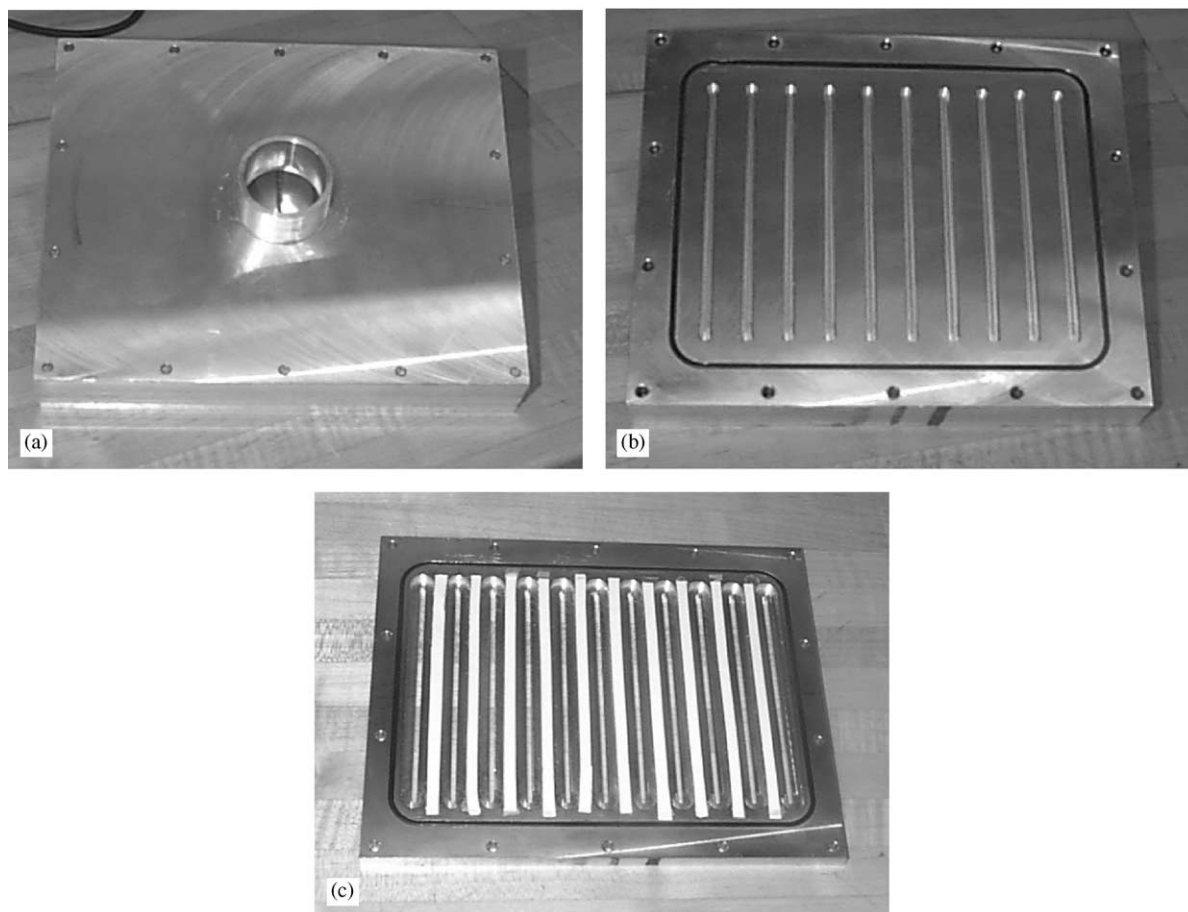


Fig. 1. Photographs of the high volume, low pressure drop ultrafine PM separator: (a) inlet; (b) acceleration slit nozzle block; and (c) collection substrate block.

factor, given by the following equation (Hinds, 1982):

$$C_c = 1 + \frac{2}{Pd_p} [6.32 + 2.01 \exp(-0.1095Pd_p)], \quad (2)$$

where P is the absolute pressure in the impaction region (in cmHg) and d_p is the particle diameter in μm .

2.2. Laboratory tests: effect of impaction substrate

To obtain accurate particle separation and classification with any impactor, all particles colliding on the impaction surface must adhere to it. One of the most common errors in the size distribution measurements obtained by means of impactors is introduced when dry solid particles bounce from previous stages and are collected in a subsequent stage or an after-filter. Particle bounce depends upon the nature of the impaction surface, the type and thickness of coating material, the type of

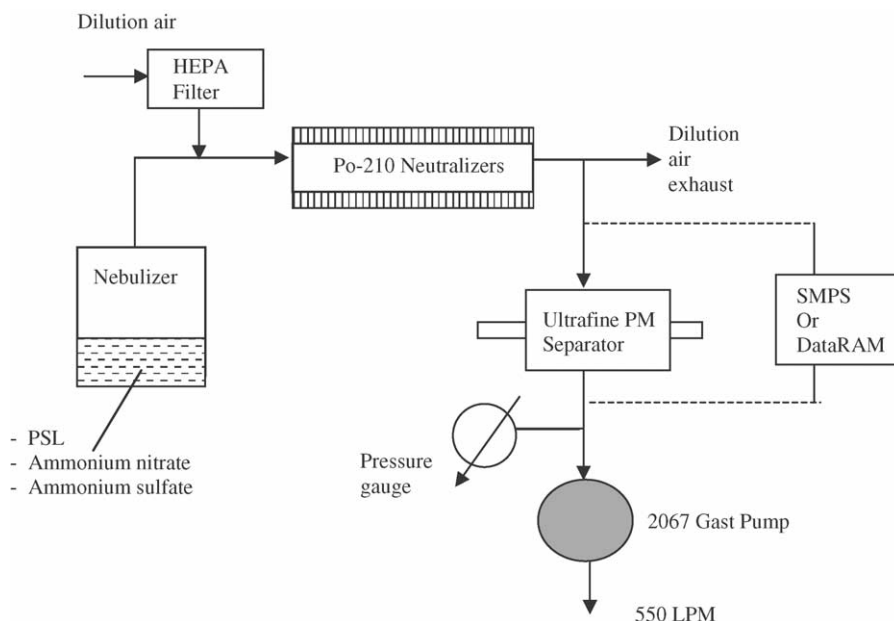


Fig. 2. Schematic of the experimental setup used for the laboratory characterization of the ultrafine PM separator.

particles, particle loading on the impaction surface, the sampling conditions and the design of the impaction substrate (Rao & Whitby, 1978a, b; Turner & Hering, 1987; Tsai & Cheng, 1995). Some investigations have indicated that an impaction substrate coated with an adhesive material can reduce particle bounce and reentrainment at light particles loading. However, uncoated surfaces are highly desirable, as they enable chemical and elemental analysis of the collected particulate matter that is free of interferences from coating materials. They are also more practical in high-temperature sampling conditions (Biswas & Flagan, 1988).

The use of an uncoated substrate as the impaction surface was considered a highly desirable feature in the design of our impactor. As part of our experimental characterization, we therefore investigated the possibility of achieving high particle collection efficiency on uncoated substrate surfaces. Three different substrates were tested, viz., a double-sided tape (3M Corp, St. Paul, MN), a quartz filter substrate (Tissuquartz, Pall Gelman, Ann Arbor, MI), and the original uncoated aluminum surface of the substrate. Particle collection efficiency for each substrate type was plotted against particle aerodynamic diameter to identify an optimum configuration that would maximize particle collection efficiency without the use of an adhesive coating.

The experimental setup for testing the effects of substrate surface type and material is shown schematically in Fig. 2. For particles in the range of 0.025–0.50 μm , polydisperse aerosols were generated by atomizing dilute aqueous solutions of ammonium sulfate and ammonium nitrate, using a constant output nebulizer (HEART, VORTRAN Medical Technology Inc., Sacramento, CA). The generated particles were drawn through a 2-l glass container in which they were mixed with room dry air in order to remove excess moisture. The dry aerosols were then drawn through a series of Po-210 neutralizers (NDR Inc., Grand Island, NY) that reduced particle charges before entering the USC ultrafine separator. Penetration was determined by measuring the particle count concentrations

for different sizes upstream and downstream of the USC ultrafine separator using a Scanning Mobility Particle Sizer (SMPS Model 3936, TSI Inc., St. Paul, MN). For each particle size, particle penetration (or 1—collection efficiency) was defined as the ratio of the concentration measured downstream to that measured upstream of the impactor. In addition to the laboratory-generated ammonium nitrate and sulfate aerosols, indoor air particles in the size range of 0.025–0.3 μm were also used as the test aerosol. Particle penetration through the USC ultrafine separator was determined again by measuring their concentrations upstream and downstream of the impactor by means of the SMPS.

A similar experimental setup was used for particles in the range of 0.3–2.5 μm . Monodisperse aerosols were generated by atomizing dilute aqueous suspensions of polystyrene latex particles (PSL, Bangs Laboratories Inc., Fisher, IN). Particle penetration through the impactor was measured as a function of particle size by means of a nephelometer, DataRAM (RAM-1, MIE Inc., Billerica, MA), which was used to measure the mass concentrations of the monodisperse upstream and downstream of the ultrafine separator. The upstream and downstream measurements were repeated at least three times. The contributions from background ambient concentrations before and after the USC ultrafine PM separator were recorded and subtracted from those of the input and concentrated aerosols prior to determining the collection efficiencies at the given particle size. It should be noted that indoor air levels were on the order of 7–12 $\mu\text{g m}^{-3}$, and substantially smaller than those of the generated aerosols (prior to entering the USC ultrafine impactor), which varied from 220 to about 500 $\mu\text{g m}^{-3}$. Therefore, the contributions of the indoor aerosol to the overall concentrations measured upstream and downstream of the USC ultrafine impactor were considered negligible.

2.3. Field tests

In addition to the laboratory experiments, the performance of the USC ultrafine separator was evaluated in limited field experiments conducted in the environment of the University of Southern California, approximately 3 km south of downtown Los Angeles. For these field tests, the impactor was used in conjunction with the MSP HVVI Model 340, which was placed upstream of its inlet to separate coarse PM (2.5–10 μm) from the air sample entering the USC impactor. The USC impactor was connected to the major flow of the HVVI, which operated with half of its virtual impactors blocked. Quartz strips cut from 20 \times 25.4 cm^2 quartz filter sheets (Pallflex Corp., Putnam, CT) were used as impaction substrate in the USC ultrafine separator, as they turned out to be the most efficient substrate based on the laboratory characterization tests. Each strip was 12.5 \times 0.5 cm^2 . A 47 mm quartz filter (Pall Gelman, Ann Arbor, MI), connected to a small pump sampling at 30 l min^{-1} was placed immediately downstream of the USC ultrafine separator to sample ultrafine particles penetrating the impactor, whereas the remaining 520 l min^{-1} of the air flow was drawn by the Gast 2067 pump. A collocated Micro orifice Uniform Deposit Impactor (MOUDI Model 110, MSP Corp., Minneapolis, MN) was used as a reference sampler, sampling at a flow rate of 30 l min^{-1} . For our tests, three of the MOUDI stages were used classifying particles in the following size intervals: $\geq 2.5 \mu\text{m}$, 0.16–2.5 μm and a back up filter collecting PM < 0.16 μm . The 2.5 μm cut MOUDI stage was coated with Silicon lubricant (Chemplex 710, NFO Technologies, Kansas City, KS) to minimize particle bounce. A prebaked 47 mm aluminum foil disk was used for the stage collecting particles between 0.16 and 2.5 μm , while a 37 mm prebaked quartz filter (Pall Gelman), on the back up, collected PM < 0.16 μm . These substrates were chosen because they

can be analyzed conveniently for both elemental (EC) and organic carbon (OC) and for inorganic ions.

The sampling flow of the small pump used downstream of the USC impactor was purposely chosen to match that of the MOUDI in order to ensure that potential sampling biases due to either adsorption of organic vapors on the quartz filter, or to volatilization of ammonium nitrate and semivolatile organic compounds would be similar for both samplers. Sampling of ambient aerosols in each of the field tests lasted for around 10–12 h. A total of eight field tests were performed from April 3, 2001 to April 16, 2001.

At the end of each field experiment, the aluminum foil disks and the quartz filters of the MOUDI and the ultrafine impactor were carefully cut in half. One half of each filter was analyzed by means of ion chromatography to determine the concentrations of particulate sulfate and nitrate ions. The other half was analyzed by means of thermoanalysis to determine the EC and OC content of accumulation and ultrafine PM. A slice of approximately 0.2 cm^2 from each filter was placed in a platinum boat containing MnO_2 . The sample was acidified with an aliquot of HCl and heated to 115°C to dehydrate the sample, and form CO_2 as an index of particle-associated carbon. The boat was then inserted into a dual zone furnace, where MnO_2 oxidized OC at 550°C and EC at 850°C . A Flame Ionization Detector (FID) converted the CO_2 combustion product to CH_4 for detection. This analytical method is more elaborately described by Fung (1990). Similarly, half of the quartz strips of the USC ultrafine impactor in each field experiment were analyzed by means of ion chromatography and half by thermo-analysis, as described above.

3. Results and discussion

3.1. Laboratory characterization of the USC ultrafine PM separator

Results from the laboratory characterization of the USC ultrafine separator are shown in Figs. 3 and 4. Fig. 3 shows the plot of the impactor's collection efficiency as a function of particle size for various substrate surfaces used. Polydisperse ammonium sulfate aerosols were used for particle range between 0.025 and $0.3\text{ }\mu\text{m}$, while monodisperse PSL particles were used for $0.49\text{--}2.6\text{ }\mu\text{m}$ range. The plotted collection efficiency data correspond to averages of three repeated runs per particle size and impaction substrate (the observed variability in each test was less than $\pm 10\%$). The data plotted in Fig. 3 clearly illustrate the superiority of the quartz substrate used as the impaction surface. Particle collection efficiency increases rapidly with particle size for all three substrates. For any particle size, the collection efficiency obtained for quartz substrates is higher than those obtained for the other two surfaces. The increased collection efficiency observed for smaller particles could be attributed to partial entrainment of the impinging air streamlines into the upper layers of the quartz substrate fibers, which would tend to cause capture of particles somewhat smaller than the impactor's actual cutpoint. Similar observations were made by Willeke (1975) in experiments performed to characterize a high-volume slotted cascade impactor (Model 235, Sierra Instruments, Atlanta, GA). Aluminum, and 3M double-sided tape surfaces are much harder compared to quartz fiber, thus less penetration of the impinging jet occurs for these surfaces.

For particles larger than about $0.2\text{ }\mu\text{m}$, the very high collection efficiency observed for the quartz substrates is clearly due to the elimination of particle bounce, which seems to be more pronounced

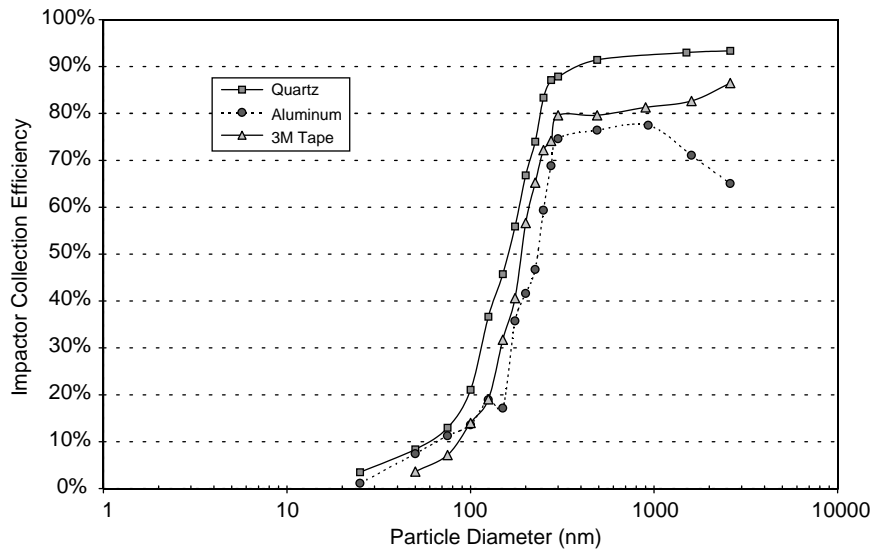


Fig. 3. Evaluation of the USC high volume low cutpoint impactor with different surfaces as impaction substrates.

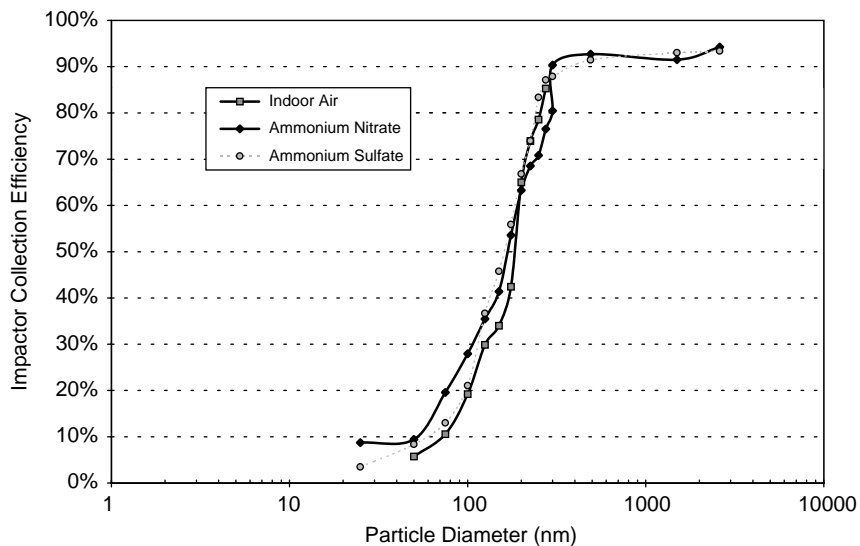


Fig. 4. Evaluation of the USC high volume low cutpoint impactor with an uncoated quartz substrate and different types of test aerosols.

for the uncoated aluminum substrate, possibly because this substrate is a harder surface than quartz or 3M tape. The results shown in Fig. 3 indicate that particle collection efficiency for the quartz substrate increases to more than 90% as particles size becomes larger than approximately $0.2\ \mu\text{m}$. By contrast, the collection efficiency for the 3M tape reaches 80% for particles of $0.2\ \mu\text{m}$, whereas the efficiency for the aluminum substrate never exceeds 75% and falls sharply with particle size for

particles larger than 0.8 μm . The decreased collection efficiency observed for the double-sided 3M tape is somewhat surprising, considering that this tape is partially coated with a very thin adhesive layer. This result indicates that apparently this coating is not sufficient to prevent particle bounce at jet velocities on the order of 5500–6000 cm s^{-1} (such as those of the USC ultrafine separator) or higher.

The experimental data plotted in Fig. 3 show that the 50% cutpoint of the USC ultrafine impactor is approximately at 0.15 μm , corresponding to a \sqrt{St} of 0.42. This is a somewhat lower \sqrt{St} value than those typically found in rectangular geometry impactors, ranging from 0.58 to 0.75 (Willeke, 1975; Marple & Rubow, 1986; Hering, Gundel, & Daisey, 1997). Most of these experimentally determined \sqrt{St} values were obtained for rectangular impactors using aluminum or some other type of a coated metal surface acting as the impaction substrate. The smaller \sqrt{St} values obtained in this study are most likely due to the penetration and capture of particles smaller than the impactor's cutpoint within the front layers of the quartz fiber substrate. This mechanism is responsible for eliminating bounce and reentrainment of particles collected on the quartz strips. Similar observations were made in a low cutpoint impactor using polyurethane foam (PUF) as the impaction substrate (Kavouras, Ferguson, Wolson, & Koutrakis, 2000). The authors of that study similarly attributed the higher collection efficiency of smaller than the theoretical cutpoint particles to partial entrainment of these particles within the PUF. Apparently, porous (PUF) or fibrous (quartz) materials used as impaction substrates result in decreasing the 50% cutpoint of the impactor as well as reducing (if not eliminating) particle bounce, both of which are highly desirable attributes of an impaction surface.

The sharpness of the collection efficiency curve of an impactor can be defined in terms of the geometric standard deviation (σ_g), which is the square root of ratio of the aerodynamic particle diameter corresponding to 84% collection efficiency to that corresponding to 16% efficiency (Marple & Willeke, 1976). Based on this definition, the value of σ_g is approximately 1.3–1.4 for this impactor when the quartz substrate is used, thereby indicating reasonably sharp aerodynamic particle separation characteristics.

Fig. 4 shows a plot of the particle collection efficiency as a function of particle size using different types of test aerosols and the uncoated quartz filter strips as the impaction substrate. The quartz surface was used because it was found to be the optimum impaction substrate of all three surfaces tried. The inlet relative humidity in these experiments varied from 20% to 45%. The data plotted in Fig. 4 indicate that the particle collection efficiency obtained for ammonium nitrate particles is practically identical to that observed for non-volatile ammonium sulfate and the real-life indoor air particles, which comprise mostly of hydrophobic compounds such as organic and elemental carbon. These results suggest that particle properties such as hygroscopicity and volatility do not seem to affect the degree to which these particles would be captured by the impactor, a finding that is not surprising given that particle separation and capture by the impactor occurs under near-atmospheric pressure.

3.2. Particle loading capacity tests

One of the main concerns in conventional impactors is the possible degradation of the collection efficiency with particle loading. Previous investigations on the effect of loading on the impactor's performance characteristics showed that the collection efficiency of impactors using coated substrates degrades over time because the grease becomes ineffective with particle accumulation (Reischl &

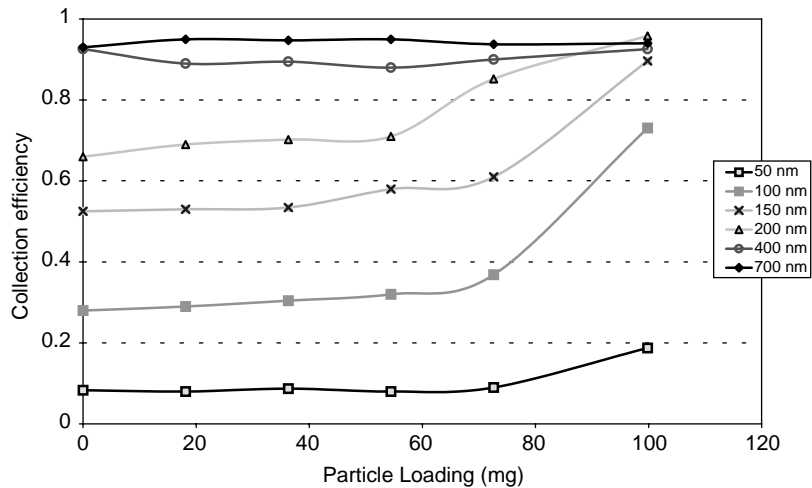


Fig. 5. Collection efficiency as a function of particle loading for different particle sizes.

John, 1978; Turner & Hering, 1987; Tsai & Cheng, 1995). Incoming particles bounce off those previously deposited. This degradation poses a serious limitation to sampling high concentration aerosols or sampling over prolonged time periods, both of which would lead to high particle loadings.

The effect of particle loading on the performance of the USC ultrafine separator was investigated by generating polydisperse ammonium sulfate aerosol in a process similar to that used to characterize the impactor's collection efficiency. The mass concentration of the generated aerosol was determined by means of a 47-mm filter (PTFE Teflon, 2 μm pore, Gelman Science Ann Arbor, MI) sampling at a flow rate of 30 l min^{-1} and in parallel to the USC ultrafine particle separator. The volume median diameter of the generated aerosol, measured by means of the SMPS, was approximately $0.35\text{ }\mu\text{m}$. SMPS measurements upstream and downstream of the impactor in the beginning of this experiment further confirmed that over 95% of the aerosol by mass is collected by the impactor. The stability of the concentration of the generated aerosol throughout the loading test was confirmed by means of the DataRAM, also sampling in parallel to the USC ultrafine separator, SMPS and Teflon filter samplers. Every 10 min, particle penetration through the impactor was determined for selected particle sizes from 0.05 to $0.7\text{ }\mu\text{m}$ by adjusting manually the voltage of the SMPS and by measuring the concentrations for each size upstream and immediately downstream of the impactor. At the end of the experiment, which lasted for about 1 h, the Teflon filter was weighed using a Mettler 5 Microbalance (MT 5, Mettler-Toledo Inc., Highstown, NJ), under controlled relative humidity (e.g. 40–45%) and temperature (e.g., $22\text{--}24^{\circ}\text{C}$) conditions. The mass concentration of the ammonium sulfate test aerosol was 3.3 mg m^{-3} resulting in a total impactor loading of approximately 108 mg in 1 h.

Results from the loading tests are shown in Fig. 5, in which the collection efficiency of the impactor for six different particle sizes varying from 50 to 700 nm is plotted as a function of particle loading, expressed in mg of accumulated particles. The data plotted in Fig. 5 clearly demonstrate that particle bounce and reentrainment, which would have been manifested by a detectable decrease in the collection efficiency measured for any size, do not occur for loadings at least as high as

60–70 mg. In fact, the data of Fig. 5 indicate that the impactor's efficiency increases with loading. This increase is particularly pronounced at loadings exceeding approximately 60 mg and for particles in the 0.1–0.2 μm range, thus close to the 50% cutpoint of the impactor. The increase in small particle capture with high particle loadings is probably due to the decrease in the gap between the impactor's jet and the collection substrate caused by the excessive accumulation of particulate matter on the substrate. This conclusion can be further supported by the increase in pressure drop across the impactor observed after about 45 min (or about 75 mg) of loading. The pressure drop at that time increased abruptly from its normal value of 0.02 to 0.045 kPa. This increase in pressure drop is highly undesirable, as it will ultimately affect the stability of the operation of the impactor by potentially leading to rapid clogging. The results plotted in Fig. 5, however, indicate that the performance of the impactor is very stable at least up to about 60 mg of particle loading. For this loading, the average thickness of deposit could be estimated based on the total area of deposit, particle density (1.78 g cm^{-3} for ammonium sulfate) and the packing density of the PM deposit, which is assumed to be between 0.5 and 0.6 (Tsai & Cheng, 1995). The area of the deposit is estimated assuming that the length of each deposit associated with an acceleration nozzle has the same length with the nozzle and is about 1.5–2 times wider (Zhang & McMurry, 1987). Based on these assumptions, the total particulate deposit area is approximately $2.8\text{--}3.8 \text{ cm}^2$ and the thickness of the deposit is between 0.018 and 0.023 cm, which is much smaller than the distance of 0.07 cm between the acceleration nozzle and the impaction substrate and should not lead to an increase in pressure drop across the impactor. Assuming a $\text{PM}_{2.5}$ concentration of $40 \mu\text{g m}^{-3}$, the impactor can operate uninterruptedly for about 46 h. Since human exposures to concentrated PM last for about 2 h, these results imply that the quartz impaction substrates will need to be replaced after approximately 20 exposures. Lower ambient concentration levels would prolong further the use of these substrates before replacement is necessary.

3.3. Field evaluation of the USC ultrafine PM separator

Results from the field evaluation of the USC ultrafine separator are summarized in Figs. 6–9. In each figure, the ultrafine or accumulation PM mode concentrations determined by the USC impactor are compared to those determined by the MOUDI. The coordinates are fit by a linear regression and the tightness-of-fit by correlation coefficients. The slopes of the regression lines thus provide an average estimate of the USC impactor-to-MOUDI concentration ratio, hence a measure of the overall agreement obtained between these two samplers. MOUDI and USC impactor concentrations appear to be highly correlated, with correlation coefficients (R^2) ranging from 0.87 to 0.93.

The nitrate and sulfate accumulation mode concentrations of the USC and MOUDI impactors agree within 5%, as indicated by the data shown in Fig. 6. Comparisons between the ultrafine PM concentrations measured by the MOUDI and USC impactors based on inorganic ions were only possible for sulfate, as the ultrafine nitrate concentrations measured by the MOUDI were below the limit of detection of ion chromatography. This may be due to either the very low nitrate concentration of ultrafine PM, and/or to volatilization losses of ammonium nitrate from ultrafine particles, given that these particles are collected by the MOUDI after-filter under reduced pressure of 0.63 atm. A recent study by Chang, Sioutas, Kim, Gong, and Linn (2000) led to similar findings regarding loss of nitrate from ultrafine PM collected in the MOUDI after-filter. The agreement between the MOUDI and USC impactors based on ultrafine PM sulfate, however, is excellent, with the USC concentrations

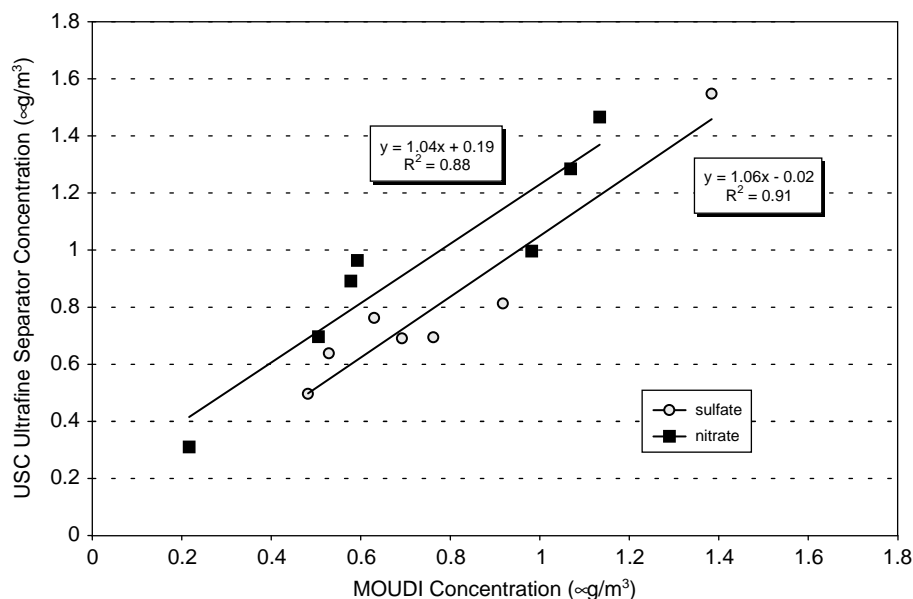


Fig. 6. Accumulation mode PM sulfate and nitrate concentrations obtained by means of the MOUDI and the USC ultrafine PM separator.

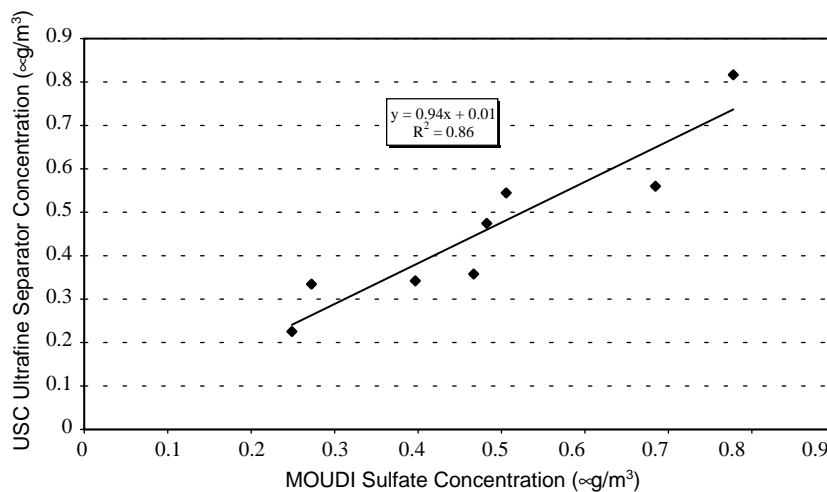


Fig. 7. Ultrafine PM sulfate concentrations obtained by means of the MOUDI and the USC ultrafine PM separator.

being about $0.94 (\pm 0.07)$ those measured by the MOUDI. MOUDI and USC impactor concentrations are also highly correlated ($R^2 = 0.87$), as shown in Fig. 7.

Similar to the sulfate concentrations, the ultrafine PM elemental and organic carbon concentrations obtained by means of the MOUDI and the USC impactor are in excellent agreement, with the average USC-to-MOUDI concentrations being $0.90 (\pm 0.06)$ and $1.10 (\pm 0.09)$ for OC and EC, respectively. The data are also highly correlated, with R^2 of 0.87 and 0.92 for EC and OC,

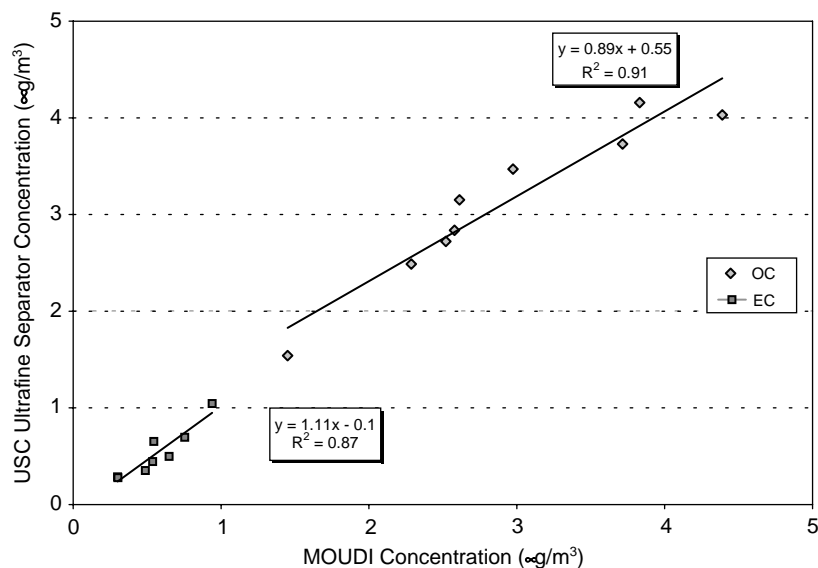


Fig. 8. Ultrafine PM elemental carbon (EC) and organic carbon (OC) concentrations obtained by means of the MOUDI and USC ultrafine separator.

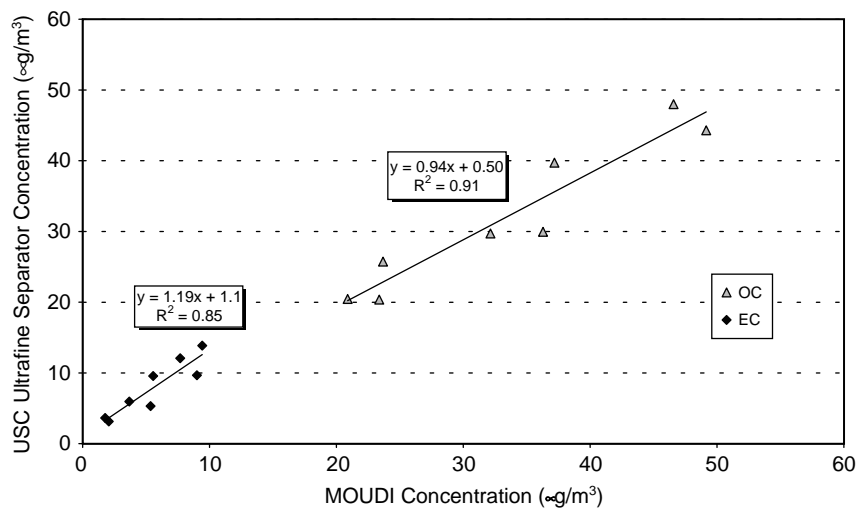


Fig. 9. Accumulation mode PM elemental carbon (EC) and organic carbon (OC) concentrations obtained by means of the MOUDI and the USC ultrafine PM separator.

respectively (Fig. 8). Excellent agreement was also observed between the accumulation PM mode OC concentrations measured by the MOUDI and USC impactors, with overall sampler agreement being within 6% (Fig. 9). The accumulation mode EC concentrations of the USC impactor were found to be slightly higher than those measured by the MOUDI, with the average USC impactor-to-MOUDI ratio being $1.19 (\pm 0.11)$. The high degree of correlation ($R^2 = 0.85$) between the accumulation

mode EC data suggests that, although small, this difference between the two samplers might be systematic. One possible explanation for the slightly higher EC concentrations of the USC impactor might be due to the differences in the shape of the collection efficiency curves of the MOUDI and USC impactor stages collecting the accumulation mode particles. The MOUDI collection efficiency curves are generally very steep, with typical geometric standard deviation (σ_g) values on the order of 1.1–1.2 (Marple, Rubow, & Behm, 1991), compared to 1.4 of the USC impactor (Fig. 4). This difference is primarily due to differences in the air flow patterns between round and rectangular jet impactors, with the round jet impactors resulting always in steeper collection efficiency curves (Marple & Rubow, 1986). The relatively higher capture of particles smaller than $0.15\text{ }\mu\text{m}$ by the USC impactor may result in somewhat higher EC concentrations measured in the accumulation mode, as ultrafine particles are generally rich in EC. Overall, the results plotted in Figs. 6–9 indicate that, despite the relatively small number of field tests, the accumulation and ultrafine PM mode concentrations obtained by the two samplers are in practically excellent agreement.

4. Summary and conclusions

A high-volume, low pressure drop impactor for separation of ultrafine from accumulation mode particles has been developed and evaluated. The impactor operates at 550 l min^{-1} with a pressure drop of 0.020 kPa and has a 50% cutpoint of $0.15\text{ }\mu\text{m}$ in aerodynamic diameter.

Laboratory characterization was performed using ammonium sulfate and nitrate as well as PSL particles. These experiments corroborated the 50% cutpoint of the impactor at $0.15\text{ }\mu\text{m}$. The tests further identified quartz fiber substrates as an optimum impaction surface, resulting in collection efficiencies reaching above 90% for particles larger than approximately $0.2\text{ }\mu\text{m}$ without the use of an adhesive coating.

Particle loading tests were conducted by generating polydisperse ammonium sulfate aerosols at concentrations of 3.3 mg m^{-3} , resulting in a total impactor loading of 108 mg in 1 h . No detectable loss in the impactor's collection efficiency was observed for particle loadings up to 60 mg . However, for loadings higher than $60\text{--}70\text{ mg}$, the impactor's collection efficiency for particles in the $0.1\text{--}0.2\text{ }\mu\text{m}$ range as well as the pressure drop across its jets were both sharply increased. These two phenomena may be due to the decrease in the gap between the impactor's jet and the collection substrate caused by the excessive accumulation of particulate matter on the substrate. These experiments therefore identified 60 mg as a maximum loading level beyond which the impactor's operation will likely become unstable due to clogging.

A field comparison between the USC ultrafine separator and MOUDI conducted outdoors showed excellent overall agreement between the accumulation and ultrafine PM mode concentrations of nitrate, sulfate, OC and EC obtained by the two samplers. MOUDI and USC impactor concentrations were highly correlated, with R^2 ranging from 0.87 to 0.93. The nitrate and sulfate accumulation mode concentrations of the USC and MOUDI impactors agreed within 5%. The agreement between the MOUDI and USC impactors based on ultrafine PM sulfate was excellent, with the USC concentrations being about $0.94 (\pm 0.07)$ those measured by the MOUDI. Similar to the sulfate concentrations, the ultrafine PM elemental and organic carbon concentrations obtained by means of the MOUDI and the USC impactor are in excellent agreement, with the average USC-to-MOUDI concentrations being

0.90 (± 0.06) and 1.10 (± 0.09) for OC and EC, respectively. Excellent agreement was also observed between the accumulation PM mode organic carbon concentrations measured by the MOUDI and USC impactors, with overall sampler agreement being within 6%.

This impactor has been developed primarily as a separator of ultrafine from accumulation mode particles for use in human exposure studies to concentrated ambient ultrafine aerosols. Ultrafine particles penetrating the impactor can either be directly supplied to exposure chambers, or collected on standard $20 \times 25.4 \text{ cm}^2$ quartz or Teflon filters. High-volume collection of size-fractionated PM accomplished by this impactor further enables investigators in the field of environmental health to conduct toxicological studies using ambient accumulation and ultrafine mode particles in vitro as well as by means of intratracheal instillation.

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